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(54) Packaging bags.

(57) A packaging bag has an outer layer and an inner layer. The outer layer is a low density ethylene-based polymer or a mixture thereof with an ethylene-vinyl acetate copolymer. The inner layer is a mixture of a high density polyethylene and an ethylene-α-olefin copolymer. This packaging bag has excellent physical properties, e.g. tensile strength and impact resistance, and is suitable for use in the packaging of heavy materials, particularly by an automatic packaging technique.

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TITLE:

Packaging Bags

DESCRIPTION:

The invention relates to packaging bags, and more particularly, to packaging bags which have high stiffness and blocking resistance and are therefore suitable for automatic packaging.

Heretofore, heavy-duty bags for packaging granular materials such as rice and wheat, powdery materials such as fertilizer and feed, and angular solid materials such as fowl have been made of low density polyethylene. These heavy-duty bags, however, are limited in their usefulness because of the low mechanical strength of low density polyethylene. In order to compensate for the poor mechanical strength, therefore, it is necessary to increase the film thickness. Particularly, in the case of heavy-duty bags, it is required for the film thickness to be markedly increased because of their seriously low tensile strength and stiffness. Furthermore, low

density polyethylene is not suitable for use in the production of bags for automatic packaging because its stiffness is poor.

In recent years, therefore, bags made of a laminated film comprising a high density polyethylene layer and a low density polyethylene layer have been proposed to overcome the above-described problems of conventional bags (see Japanese Patent Application Laid-Open Nos 92023/81, 29426/82, and 30994/80).

A bag prepared by using a film as disclosed in Japanese Patent Application Laid-Open Nos. 92023/81 and 29426/82 is very preferred for practical use because the seal strength and the film impact strength are high, the tear strength is well balanced, and furthermore, the outer layer is not slippery. However, when the bag is used to accommodate and convey angular heavy materials such as fowl, its film puncture strength, tear strength, impact resistance at low temperatures, and so forth become problems. Also, a bag made of a film as disclosed in Japanese Patent Application Laid-Open No. 30994/80 suffers from disadvantages in that since the inner layer of the bag is made of low density polyethylene, blocking readily occurs and difficulties are encountered in opening the bag and, furthermore, it takes much labour and time

to place materials in the bag because the inner surface is slippery.

The invention provides a packaging bag comprising an outer layer and an inner layer, characterised in that the outer layer is a low density ethylene-based polymer or a mixture thereof with an ethylene-vinyl acetate copolymer, and the inner layer is a mixture of a high density polyethylene and an ethylene-α-olefin copolymer.

The outer layer of the bag of the invention is made of a low density ethylene-based polymer (hereinafter referred to as "LDPE") or a mixture of LDPE and an ethylene-vinyl acetate copolymer (hereinafter referred to as "EVA"). Various types of LDPE can be used in the invention. Usually they have a density of from 0.91 to 0.94 gram per cubic centimeter (g/cm^3), preferably from 0.915 to 0.935 g/cm^3. Their melt indexes (MI) are within the range of from 0.3 to 10 grams per 10 minutes (g/10 min), preferably from 0.3 to 4 g/10 min. When MI is more than 10 g /10 min, the impact resistance is lowered, unsuitably for practical use. On the other hand, when MI is less than 0.3 g/10 min, processability becomes low. The LDPE used includes that produced under high pressure and that produced under low pressure. Examples of such LDPE produced under low pressure

include copolymers of ethylene and α-olefins containing from 3 to 12 carbon atoms (e.g. propylene, butene-1, pentene-1, hexene-1, 4-methylpentene-1 and nonene-1). The α-olefin unit content of the copolymers is preferably from 1 to 10% by mole.

When the outer layer of the present bag is made of a mixture of LDPE and EVA, the mixing ratio of LDPE to EVA is not critical. Usually, however, the ratio of LDPE/EVA is from 10/90 to 95/5 (by weight), with the range of from 60/40 to 90/10 (by weight) being preferred. If the proportion of EVA is excessively increased, various problems arise; for example, a bag produced by using the resulting resin mixture has acetic acid odour and, furthermore, the surface of the bag excessively loses its smoothness. Such poor surface smoothness causes blocking between bags and makes bags stick to conveying equipments, e.g. a roller and the belt of a belt conveyor, as used in automatic packaging, resulting in a serious reduction in workability.

The inner layer of the bag of the invention is made of a mixture of a high density polyethylene (hereinafter referred to as "HDPE") and an ethylene-α-olefin copolymer. Various types of HDPE can be used in the invention. usually they have a density ranging between 0.94 and 0.97

g/cm^3, with the range of from 0.945 to 0.965 g/cm^3 being preferred. Their melt indexes are within the range of from 0.01 to 1 g/10 min, preferably from 0.02 to 0.2 g/10 min. When MI is more than 1 g/10 min, the mechanical strength, e.g. tensile strength, of the resulting film is undesirably lowered. Various ethylene-α-olefin copolymers can be used in admixture with HDPE. The ethylene-α-olefin copolymer suitably has a density of from 0.910 to 0.940 g/cm^3, preferably from 0.915 to 0.935 g/cm^3, and a MI of from 0.3 to 10 g/10 min, preferably from 0.3 to 4 g/10 min. Preferred examples of α-olefins which can be used in the preparation of the copolymers are those monomers containing from 3 to 12 carbon atoms, such as propylene, butene-1, pentene-1, hexene-1, 4-methylpentene-1, octene-1 and nonene-1. The α-olefin unit content of the copolymer is suitably within the range of from 1 to 10% by mole.

Of these ethylene-α-olefin copolymers, ethylene-propylene-based copolymer elastomers (hereinafter referred to as "EP") are most suitable. Various types of EP can be used in the invention. Those elastomers having a Mooney viscosity of from 40 to 150 are preferred. When the Mooney viscosity is less than 40, the mechanical strength of the resulting film is undesirably lowered. Preferred examples of EP are ethylene-propylene rubber and ethylene-propylene terpolymers containing, as a third component, dienes such as divinylbenzene, 5-

-ethylidene-2-norbornene, 1, 4,-hexadiene, dicyclopentadiene and cyclooctadiene.

The ratio of HDPE to the ethylene-α-olefin copolymer is not critical and can be determined appropriately depending on the purpose for which the ultimate bag is used. Usually, however, the ratio of HDPE to the ethylene-α-olefin copolymer is from 95/5 to 50/50 (by weight), with the range of from 90/10 to 60/40 (by weight) being preferred. When the proportion of the ethylene-α-olefin copolymer is less than 5% by weight, the puncture strength and seal strength of the resulting film are reduced, and when a bag prepared by using the film is dropped, it is easily torn. On the other hand, when it is more than 50% by weight, processability becomes low, and a bag prepared by using the resulting film has poor stiffness and is not suitable for practical use.

The bag of the invention comprises the above-described outer and inner layers which are laminated on each other. Although the ratio of the thickness of the outer layer to the thickness of the inner layer can be determined depending on the purpose for which the bag is used, it is usually determined within the range of 1/99

to 60/40. When the thickness of the outer layer is more than 60% of the total thickness of the outer and inner layers, the tensile strength and stiffness of the bag are undesirably lowered. On the other hand, when it is less than 1%, the processability thereof becomes difficult.

The bag of the invention is usually of the two layer laminated structure consisting of an outer layer and an inner layer. If desired, a three layer laminated structure, including an intermediate layer between the outer and inner layers or having an innermost layer on the inner layer, can be used. The type of the intermediate layer being interposed between the outer and inner layers is not critical and can be determined appropriately depending on the purpose for which the bag is used. Although the type of the innermost layer is not critical as in the case with the intermediate layer, it is preferably made of an ethylene-α-olefin copolymer. Although the properties of the ethylene-α-olefin copolymer are not critical, the density is usually from 0.91 to 0.94 g/cm^3 and preferably from 0.915 to 0.935 g/cm^3, and MI is from 0.1 to 20 g/10 min and preferably from 0.2 to 10 g/10 min. The α-olefin content of the copolymer is usually within the range of from 1 to 10% by mole.

The bag of the invention can be produced by various techniques. For example, feed resin for the outer layer and feed resin for the inner layer are melted and kneaded at usual processing temperature in respective extruders and extruded therefrom, and then both of the resulting extruded resins are introduced into a circular die having double slit to form two layers which are bonded to each other inside the die. Thereafter, the inflation processing of the bonded two layers is performed at a blow ratio of from 2.5 to 8, preferably from 3 to 6, to obtain a tubular two layer film from which the bag of the present invention is produced. The outer layer and inner layer can be bonded together either outside or inside the die. It is preferred, however to bond together the layers inside the die since the bonding strength between the layers can be increased.

In producing the bag of the invention, if desired, additives such as pigment, a slip agent, an antioxidant, an antistatic agent and a weather resistance-improving agent may be incorporated into any one or both of the inner and outer layers at any processing step.

The bag of the invention having the above described structure is very superior in physical properties such as film puncture strength and tear strength to the

conventional bags and, furthermore, its seal strength and impact resistance at low temperature are very high. Thus the bag of the invention can be used satisfactorily to accommodate therein and transfer heavy materials, particularly heavy angular materials such as fowl. Moreover, the bag of the invention is reduced in blocking properties and has high stiffness and, therefore, it is suitable for use in automatic packaging and greatly accelerates packaging efficiency. Furthermore, since the mechanical strength of the bag is great, the thickness of the bag is sufficient to be about 10 to 200 microns and, therefore, a reduction in thickness can be achieved, which is economically advantageous.

The invention is illustrated by the following Examples. In these Examples, physical tests were carried out according to the following methods.

Impact Resistance: Measured using a film impact tester (manufactured by Toyo Seiki Seisakujo Co. Ltd; specified impact load: 30 kg.cm; impact hammer diameter 25.4 mm)
Puncture Strength: Measured according to JIS-Z-8401.
Tear Strength: Measured according to JIS-Z-1702.
Tensile Modulus: Measured according to JIS-Z-1702.
Tensile Strength: Measured according to JIS-Z-1702.
Elongation: Measured according to JIS-Z-1702.

Seal Strength: Measured according to ASTM-D-1822.

Practical Drop Test: A bag was charged with 20 kg of chemical fertilizer and dropped on a concrete floor from a height of 2.5 meters in such a manner that the broad bag surface was horizontal to the floor. The bag--breakage ratio (number of broken bags/number of dropped bags) was determined.

Processability: By observing the continuous operation condition in processing a two layer film by an inflation method, the processability was evaluated on a numeric scale as follows:

3 ... Excellent, i.e. continuous operation can be performed stably for more than one week.

2 ... Good, i.e. continuous operation can be performed for 1 to 6 days.

1 ... Fair, i.e. continuous operation can be performed for 3 to 24 hours.

Appearance: Evaluated by observing with the eye.

Blocking: Evaluated by examining whether or not the inner surfaces of a bag closely stick to each other in opening the bag after the production thereof.

3 ... The bag can be opened with ease.

2 ... The bag can be opened with slight difficulty.

EXAMPLES 1 to 16

Feed resin for an inner layer and feed resin for an outer

layer as shown in Table 1 were melted and kneaded in the respective extruders and extruded therefrom, and thereafter, both of the resulting extruded resins were introduced into a circular die having double slit to form two layers which were bonded to each other inside the die. Then, inflation processing was performed at a blow ratio of 4.3 to obtain a tubular two layer film in which the ratio in thickness of the inner layer to the outer layer was 4:1 and the total thickness was 80 microns. From this tubular film was produced a bag having a width of 500 mm and a length of 620 mm wherein one end of the bag was heat-sealed. Physical tests were carried out on the bags, and the results are shown in Table 1.

In Table 1, the plastics materials are identified by single letters as follows:

A: High-pressure-produced LDPE having a density of 0.934 g/cm^3 and a MI of 3 g/10 min.

B: High-pressure-produced LDPE having a density of 0.926 g/cm^3 and a MI of 0.4 g/10 min.

C: High-pressure-produced LDPE having a density of 0.921 g/cm^3 and a MI of 0.6 g/10 min.

D: High-pressure-produced LDPE having a density of 0.917 g/cm^3 and a MI of 8 g/10 min.

E: High-pressure-produced LDPE having a density of

924 g/cm^3 and a MI of 3 g/10 min.

F: Low-pressure-produced LDPE having a density of 0.921 g/cm^3, a MI of 3.7 g/10 min, and a C_8 α-olefin content of 10.3% by weight.

G: EVA having a density of 0.94 g/cm^3, a MI of 0.6 g/10 min, and a vinyl acetate content of 15% by weight.

H: HDPE having a density of 0.954 g/cm^3 and a MI of 0.05 g/10 min.

I: Ethylene-propylene copolymer having a Mooney viscosity ML_{1+4}(100°C) of 60, and a propylene content of 27% by weight.

J: Ethylene-propylene terpolymer having a Mooney viscosity ML_{1+4} (100°C) of 90 and a propylene content of 28% by weight.

K: Ethylene-propylene terpolymer having a Mooney viscosity ML_{1+4} (100°C) of 105 and a propylene content of 43% by weight.

L: Ethylene-propylene terpolymer having a Mooney viscosity ML_{1+4} (100°C) of 42 and a propylene content of 43% by weight.

EXAMPLES 17 and 18

The bags of Examples 3 and 4 respectively were charged with 20 kilograms (kg) each of chemical fertilizer, were allowed to stand one day and night in a room

maintained at -20°C, and then were subjected to the practical drop test to determine the bag breakage ratio. In Example 17 the bag breakage ratio was 15%, whilst the bag breakage ratio in Example 18 was 0%.

Table 1

Example	Inner Layer HDPE Type	Inner Layer HDPE wt%	Inner Layer EP Type	Inner Layer EP wt%	Outer Layer LDPE Type	Outer Layer LDPE wt%	Outer Layer EVA Type	Outer Layer EVA wt %	Strength of Bag: Impact Resistance kg.cm	Strength of Bag: Puncture Strength kg.cm	Strength of Bag: Tear Strength MD/TD kg.cm	Strength of Bag: Tensile Modulus MD/TD, kg/cm^2	Strength of Bag: Tensile Strength MD/TD, kg/cm^2	Strength of Bag: Elongation MD/TD %
1	H	95	J	5	B	100	-		13.0	13.0	22/35	7430/7400	390/400	460/510
2	H	90	J	10	B	100	-		13.0	48.0	40/80	6600/6890	380/400	540/500
3	H	80	J	20	B	100	-		13.0	96.0	67/110	4810/4960	370/360	540/500
4	H	70	J	30	B	100	-		14.0	112.0	90/110	3680/3820	350/360	570/520
5	H	60	J	40	B	100	-		15.0	148.0	120/110	3000/3200	340/330	580/550
6	H	50	J	50	B	100	-		16.0	165.0	130/110	2500/2700	330/320	580/550
7	H	80	J	20	C	100	-		12.0	90.0	50/90	5300/5600	370/430	510/460
8	H	80	J	20	A	100	-		11.0	48.0	40/70	5320/5680	370/420	500/450
9	H	80	J	20	E	100	-		11.8	48.0	45/70	5300/5700	370/425	500/460
10	H	80	J	20	D	100	-		10.0	40.0	40/60	5400/5500	370/390	510/490
11	H	80	J	20	F	100	-		11.5	60.0	50/70	5300/5800	370/400	510/490
12	H	80	K	20	A	100	-		11.5	50.0	50/70	5320/5680	370/440	500/480
13	H	80	L	20	A	100	-		11.0	54.0	60/70	5300/5700	340/390	510/500
14	H	80	I	20	A	100	-		11.0	54.0	37/71	5780/5960	360/360	560/620
15	H	80	I	20	B	100	-		13.0	60.0	60/90	5120/5330	349/330	540/440
16	H	80	J	20	A	90	G	10	11.0	60.0	45/65	5400/5560	390/430	500/440

Table 1 (continued)

Example	Seal Strength kg.cm	Practical Drop Test (Bag Breakage Ratio) (%)	Processability	Appearance	Blocking
1	2.0	40	3	good	3
2	4.0	15	3	good	3
3	>4.0	0	3	good	3
4	>4.0	0	3	good	3
5	>4.0	0	2	good	3
6	>4.0	0	1	good	3
7	4.0	0	3	good	3
8	4.0	5	3	good	3
9	4.0	5	3	good	3
10	4.0	25	2	good	3
11	4.0	0	3	good	3
12	4.0	0	3	good	3
13	4.0	0	3	good	3
14	3.5	5	3	good	3
15	3.5	5	3	good	3
16	4.0	5	2	good	3

EXAMPLES 19 to 23 .

Feed resin for an outer layer and feed resin for an inner layer as shown in Table 2 were melted and kneaded in the respective extruders and extruded therefrom, and thereafter, both of the resulting extruded resins were introduced into a circular die having double slit to form two layers which were bonded to each other inside the die. Then, inflation processing was performed at a blow ratio of 4.3 to obtain a tubular two layer structure film in which the ratio in thickness of the outer layer to the inner layer was 1:4 and the total thickness of the two layers was 50 microns. One end of the tubular two layer structure film was heat-sealed to produce a packaging bag having a width of 340 mm and a length of 570 mm.

The bag thus produced was subjected to various physical tests. The results are shown in Table 2.

In Table 2 the plastics materials are identified by single letters as follows:

M: High-pressure-produced LDPE having a density of 0.924 g/cm^3 and a MI of 3.0 g/10 min.

N: Ethylene-octene-1 copolymer having a density of 0.922 g/cm^3 and a MI of 2.3 g/10 min.

O: Low-pressure-produced HDPE having a density of 0.955 g/cm^3 and a MI of 0.05 g/10 min.

Table 2

Example	Outer Layer	Inner Layer O (wt%)	Inner Layer N (wt%)	Tensile Characteristics: Tensile Strength MD/TD (kg/cm^2)	Tensile Characteristics: Tensile Modulus MD/TD (kg/cm^2)	Tensile Characteristics: Elongation MD/TD (%)	Tear Strength MD/TD (kg/cm)	Impact Resistance (kg.cm)	Puncture Strength (kg.cm)
19	M	90	10	440/460	6000/8400	420/460	11.2/58.2	16.2	50
20	M	80	20	439/458	7220/8190	410/510	14.3/82.2	14.4	60
21	M	70	30	421/439	6180/6720	440/560	20.6/77.5	13.4	65
22	M	60	40	409/415	5210/6300	460/580	29.3/99.3	13.1	68
23	N	70	30	400/440	6200/6500	460/580	22.4/91.0	13.3	70

Table 2 continued

Example	Seal Strength (kg.cm)	Practical Drop Test (bag breakage ratio)	Blocking Resistance	Processability
19	2.0	3/10	3	3
20	3.5	0/10	3	3
21	7.0	0/10	3	3
22	7.5	0/10	3	3
23	7.0	0/10	3	3

EXAMPLES 24 and 25

Feed resin for an innermost layer, feed resin for an inner layer, and feed resin for an outer layer as shown in Table 3 were melted and kneaded in the respective extruders and extruded therefrom, and thereafter, the three resulting extruded resins were introduced into a circular die to form three layers which were bonded to each other inside the die. Then, inflation processing was performed at a blow ratio of 4.3 to obtain a 50-micron thick three layer laminated film. The laminated film thus produced was subjected to the same physical testings as in Example 1. The results are shown in Table 3.

In Table 3 the plastics materials are identified by single letters as follows:

P: Ethylene-butene-1 copolymer having a density of 0.921 g/cm^3, a MI of 3.7 g/10 min and a butene-1 content of 7.5% by weight.

Q: High-pressure-produced LDPE having a density of 0.924 g/cm^3 and a MI of 3.0 g/10 min.

R: HDPE having a density of 0.954 g/cm^3 and a MI of 0.05 g/min.

S: Ethylene-propylene rubber having a Mooney viscosity ML_{1+4} (100°C) of 60 and a propylene unit content of 27% by weight.

Table 3

Example	Innermost	Inner Layer: R (wt%)	Inner Layer: Ethylene-α-Olefin Copolymer (wt%)	Outer Layer	Ratio of Layer Thickness*	Tensile Characteristics: Tensile Strength MD/TD (kg/cm^2)	Tensile Characteristics: Tensile Modulus MD/TD (kg/cm^2)	Tensile Characteristics: Elongation MD/TD (%)
24	P	70	P 30	Q	3:6:1	355/420	6000/6500	550/600
25	P	85	S 15	P	3:6:1	350/430	5500/5800	580/600

Table 3 (continued)

Example	Tear Strength MD/TD (kg/cm)	Puncture Strength (kg.cm)	Seal Strength (kg.cm)	Blocking Resistance	Processability
24	30.0/75.0	57.5	6.0	O	O
25	35.0/60.0	60.0	7.0	O	O

* Ratio in thickness of innermost layer to inner layer to outer layer.

CLAIMS:

1. A packaging bag comprising an outer layer and an inner layer, characterised in that the outer layer is a low density ethylene-based polymer or a mixture thereof with an ethylene-vinyl acetate copolymer, and the inner layer is a mixture of a high density polyethylene and an ethylene-α-olefin.

2. A bag according to claim 1 characterised in that the low density ethylene-based polymer has a density of from 0.91 to 0.94 gram per cubic centimeter and a melt index of from 0.3 to 10 grams per 10 minutes.

3. A bag according to claim 1 or claim 2 characterised in that the low density ethylene-based polymer is an ethylene homopolymer or a copolymer of ethylene and an α-olefin containing from 3 to 12 carbon atoms.

4. A bag according to any preceding claim characterised in that the outer layer comprises from 10 to 95% by weight of the low density ethylene-based polymer and from 90 to 5% by weight of the ethylene-vinyl acetate copolymer.

5. A bag according to any preceding claim characterised in that the high density polyethylene has a density of

from 0.94 to 0.97 gram per cubic centimeter and a melt index of from 0.01 to 1 gram per 10 minutes.

6. A bag according to any preceding claim characterised in that the ethylene-α-olefin copolymer of the inner layer is an ethylene-propylene-based elastomer.

7. A bag according to any preceding claim characterised in that the inner layer comprises from 95 to 50% by weight of the high density polyethylene and from 5 to 50% by weight of the ethylene-α-olefin copolymer.

8. A bag according to any preceding claim characterised in that the ratio in thickness of the outer layer to the inner layer is from 1/99 to 60/40.

9. A bag according to any preceding claim characterised in that the bag further comprises an intermediate plastics layer between the inner and outer layers.

10. A bag according to any of claims 1 to 8 characterised in that the bag further comprises an innermost plastics layer on the face of the inner layer opposite to the outer layer.

European Patent Office

EUROPEAN SEARCH REPORT

0111602
Application number

EP 82 30 6711

DOCUMENTS CONSIDERED TO BE RELEVANT

Category	Citation of document with indication, where appropriate, of relevant passages	Relevant to claim	CLASSIFICATION OF THE APPLICATION (Int. Cl. 3)
Y	GB-A-2 028 716 (MOBIL OIL) * Claims 1-5; page 1, lines 33-52; page 2, lines 8-44; examples 1,2; tables 1,2 *	1-3,5, 8	B 32 B 27/32 B 65 D 65/40
Y	GB-A-1 368 634 (DU PONT OF CANADA) * Claims 1,5-10; page 1, line 9 - page 3, line 94 *	1,2,5, 8,10	
A	US-A-2 956 723 (L. TRITSCH) * Claims 1-4,12,13,16; column 1, lines 15-32; column 2, lines 25-39; column 4, line 74 - column 5, line 9; column 9, line 70 - column 10, line 18 *	1,2,5, 9,10	
			TECHNICAL FIELDS SEARCHED (Int. Cl. 3)
A	GB-A-1 370 355 (LEESONA) * Page 3, lines 90-97 *	4	B 32 B B 65 D
A	GB-A-2 037 660 (ASAHI-DOW LTD.) * Claims 1,5-7,13-15,23; page 6, lines 36-62; page 7, lines 3-6 *		
E	EP-A-0 069 526 (IDEMITSU PETROCHEMICAL) * Claims 1-10 *	1-10	

The present search report has been drawn up for all claims

Place of search	Date of completion of the search	Examiner
THE HAGUE	25-07-1983	BLASBAND I.

CATEGORY OF CITED DOCUMENTS

X : particularly relevant if taken alone
Y : particularly relevant if combined with another document of the same category
A : technological background
O : non-written disclosure
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